

Thermal Ionization Mass Spectrometry Uranium Results for September 2008 RFETS Waters

Prepared by: Deborah Norman, Mike Murrell, and David Janecky

Experimental Approach

Six water samples were received at LANL and processed for uranium TIMS measurements. The samples represented collections at three locations sampled December 6, 2007 and May 15, 2008. Aliquots of the samples were spiked with a ^{233}U spike, equilibrated by fuming with perchloric acid, and then chemically processed using ion-exchange columns to isolate and purify a uranium fraction. The processed samples were loaded onto triple filaments for analysis by thermal ionization mass spectrometry using a VG Sector 54 mass spectrometer. A chemistry process blank was also run that contained negligible uranium.

Results

The sample results are tabulated in Table 1. Sample locations are shown in Figure 1. A plot of $^{236}\text{U}/^{238}\text{U}$ vs. $^{235}\text{U}/^{238}\text{U}$ is shown in Figure 2. Table 2 shows calculated fractions of end-member depleted, enriched and natural uranium for this set of analyses.

Table 1. New Thermal Ionization Mass Spectrometry Uranium Results

ID	Sample ID	U (ng/g)	(+/-) (%)	238/235	(+/-) (%)	234/238 (e-6)	(+/-) (%)	236/238 (e-6)	(+/-)
8-3	RFETS GS 13 07	47.3	0.52	160.3	0.40	58.71	1.0	13.1	1.0
8-4	RFETS GS 13 08	21.5	0.25	155.2	0.18	60.60	0.30	11.9	0.4
8-5	RFETS SW093 07	9.93	0.29	143.6	0.22	66.52	0.33	4.33	0.9
8-6	RFETS SW093 08	7.11	0.25	142.9	0.20	67.30	0.50	3.34	2.9
8-7	RFETS SPP DISC 07	60.7	0.27	118.3	0.23	83.97	0.30	43.9	0.4
8-8	RFETS SPP DISC 08	58.7	0.31	117.8	0.33	84.34	0.28	44.2	0.4

Table 3. Calculated isotopic end-member contributions

Sample Locations	Sample Date	Easting	Northing	normalized to % end-member fractions		
				depleted	enriched	natural
GS13	12/6/2007	2086153.28	751870.11	28.7%	0.05%	71.3%
GS13	5/15/2008	2086153.28	751870.11	25.1%	0.05%	74.8%
SW093	12/6/2007	2085030.39	751730.14	9.1%	0.02%	90.9%
SW093	5/15/2008	2085030.39	751730.14	7.3%	0.01%	92.7%
SPP Discharge Gallery	12/6/2007	2085350.07	751764.39	58.0%	0.42%	41.6%
SPP Discharge Gallery	5/15/2008	2085350.07	751764.39	58.2%	0.42%	41.4%

Discussion

GS13 was previously sampled in the spring of 2002. Isotopic ratios among the four uranium isotopes measured have remained very similar, indicating just under 30% contribution of a depleted uranium end-member and less than 0.1% enriched uranium end-member. The December 2007 sample contained approximately 3 times the concentration of uranium compared to May of 2002, while the May of 2008 sample contained approximately 1.3 times. The large variation in uranium concentrations vs. relatively constant uranium isotopic signatures indicates dilution is a controlling factor for this location.

SW093 had not been previously sampled for uranium isotopic analysis. This sample site is up-drainage from GS13 and the SPP Discharge Gallery. In comparison to the GS13 sample analyses, uranium total concentrations are lower by more than a factor of two. Calculated isotopic contributions indicate less than 10% depleted uranium end-member and 0.2% or less enriched uranium end-member. Concentrations of uranium were decreased in the May 2008 sample, relative to the December 2007 sample, at SW093, consistent with the trend at GS13 but not to the same magnitude.

SPP Discharge Gallery samples have now been sampled for uranium isotopics four times between June of 2002 and May of 2008. This location is between surface water sampling sites SW093 and GS13. There was a shift in isotopic compositions between June 2002 and September of 2007, with the calculated contribution of depleted uranium end-member increasing from 32.6% to 56.7%. Samples from December of 2007 and May of 2008 were slightly higher in depleted uranium end-member contributions than the September of 2007 sample. Calculated contribution of enriched uranium end-member increased from 0.2% in the 2002 sample to 0.4% in 2007 and 2008 samples. Along with the shift in isotopic compositions, the concentration in samples from this site increased from 40 ng/g in the 2002 sample to 60 ng/g in 2007 and 2008 samples.

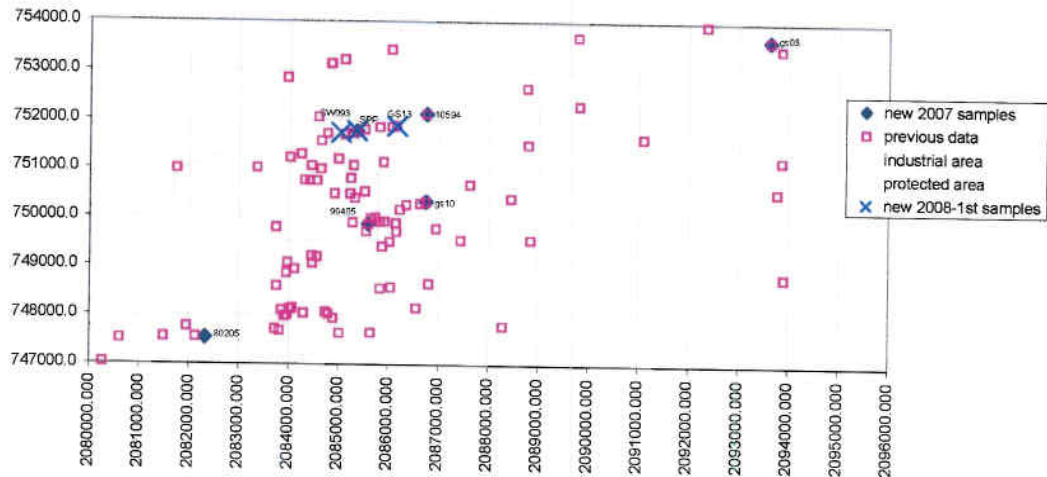


Figure 1. Sampling locations for 2008 RFETS waters.

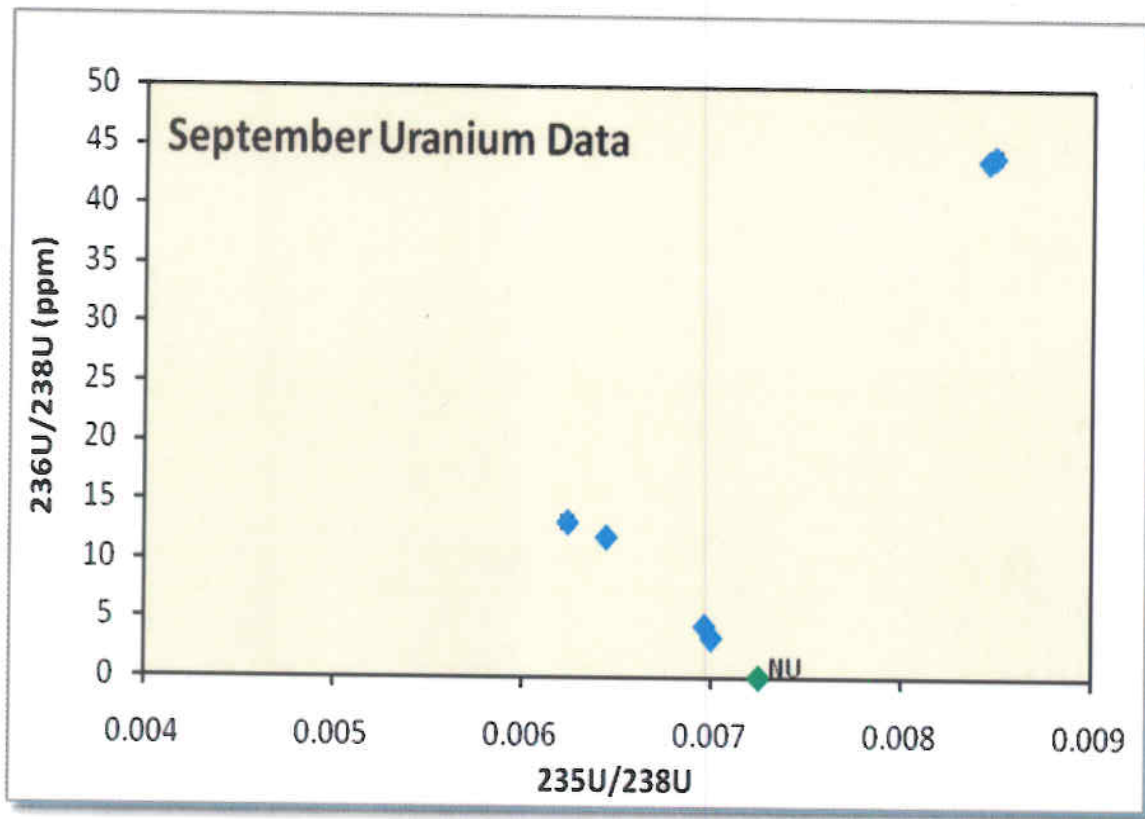


Figure 2. Plot of uranium isotopics for September analyses. None of the samples were solely natural uranium (NU). Samples to the left of the NU reference point indicate the presence of depleted uranium. Samples to the right of the NU reference point indicate the presence of the enriched uranium.